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## REMARKS

Claims 8-25 are pending in this application. By this Preliminary Amendment, Applicant AMENDS the specification and the abstract of the disclosure, CANCELS claims 1-7 and ADDS new claims 8-25.

Applicant has attached hereto a Substitute Specification in order to make corrections of minor informalities contained in the originally filed specification. Applicant's undersigned representative hereby declares and states that the Substitute Specification filed concurrently herewith does not add any new matter whatsoever to the above-identified patent application. Accordingly, entry and consideration of the Substitute Specification are respectfully requested.

The changes to the specification have been made to correct minor informalities to facilitate examination of the present application.

Applicant respectfully submits that this application is in condition for allowance. Favorable consideration and prompt allowance are respectfully solicited.

Respectfully submitted,

Date: April 6, 2005

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#### DESCRIPTION

Attorney Docket No. 36856.1340

METHOD OF PRODUCING CERAMIC MULTI-LAYER SUBSTRATE

# TECHNICAL BACKGROUND OF THE INVENTION

### 1. Field of the Invention

The present invention relates to a method of producing a ceramic multi-layer substrate on which semiconductor devices, chip capacitors, and so forthother suitable components are to be mounted.

# Background 2. Description of the Related Art

In the production of ceramic multi-layer substrates on which semiconductor devices, chip capacitors, and so forthother suitable components are to be mounted, ceramic green sheets useful for a substrate—are laminated to each other to form an unfired ceramic laminate, and then, the laminate is fired. However, in the case in whichwhen the unfired laminate is fired—as it is, the unfired ceramic laminate is shrunk atshrinks during firing, sosuch that a dimensional error is errors are caused.

Thus, for example, as described in Japanese
Unexamined Patent Application Publication No. 4243978, shrink-prevention ceramic green sheets
useful for shrink prevention, which are not sintered

at the firing temperature of the unfired ceramic laminate, are arranged on both of the main surfaces of the unfired ceramic laminate, and the unfired ceramic laminate is fired at a temperature which is higher than the sintering temperature of the unfired ceramic laminate and which is lower than the sintering temperature of the shrink-prevention ceramic green sheets—useful—for shrink prevention, and thereafter; the shrink-prevention green sheets are removed.

The specific method of removing shrinkprevention ceramic green sheets described in
W099/56510 is known, in which as a first method,
spraying ceramic powder together with compressed air,
as a second method, spraying water together with
compressed air, and as a third method, spraying a
mixture of ceramic power with water together with
compressed air are exemplifieddescribed.

However, when the first, second, and third methods are individually used, the following problems occur.

That is, according to the first method, the removing capacity is low, since the spraying spotarea of the ceramic powder is small. Also, the positional accuracy in the processing range is not high.low. Thus, possibly, the processing becomes may become unstable. As a result, it is difficult to

uniformly remove the <u>shrink-prevention</u> ceramic green sheets used for shrink-prevention. Moreover, a large-scale apparatus is required to collect the sprayed ceramic powder and the powder of the removed ceramic green sheets. Thus, <u>expensive</u> large-scale facilities are necessary, and the cost is high required.

Then, according According to the second method, a larger partportion of the shrink-prevention ceramic green sheets used can be removed. However, in the following casesituations, the shrink-prevention ceramic green sheets can not be removed. That is, if glass is contained included in the unfired ceramic laminate, the glass component of the unfired ceramic laminate and the ceramic component of the shrink-prevention ceramic green sheets will bond to each other to form a reaction layer. This reaction layer can not cannot be sufficiently be removed simply by spraying water together with compressed air.

According to the third method, the <a href="shrink-">shrink-</a>
<a href="prevention">prevention</a> green sheets can be removed more</a>
<a href="uniformly as compared with to">uniformly as compared with to</a> the method of spraying ceramic powder together with compressed air. The removal-capacity is <a href="high-increased as compared with to">high-increased as compared with to</a> the method of spraying water with compressed air. However, according to this method, <a href="fortion-to-">forto allow re-</a>
use of the spraying ceramic powder, the ceramic

powder to be sprayed is required to must have an average particle size equal to or nearly substantially equal to that of the ceramic powder of the shrink-prevention ceramic green sheets. particle size of the spraying ceramic powder is larger than that of the ceramic powder of the shrink-prevention ceramic green sheets, it will be difficult to remove the powder of the shrinkprevention ceramic sheets by means of using a filter, and hencethus, the average particle size of the spraying ceramic sheets powder changes with a lapse of use as the time that the spraying ceramic powder is used increases. As a result, the removingconditions for the shrink-prevention ceramic sheets change, and thus, it is difficult to earry out the processing—uniformly perform the processing. On the other hand, if the average particle size of the spraying ceramic sheetspowder is smaller than that of the shrink-prevention ceramic sheets, the shrinkprevention ceramic sheets can be removed by means ofusing a filter. However, in the case in which the average particle sizes of the used ceramic powders are not significantly different from each other, a partportion of the spraying ceramic powder will be also <del>be</del>removed by <del>means of</del>the filter. Thus, the average particle size of the spraying ceramic powder changes with a lapse of use as the time that the

spraying ceramic powder is used increases. As a result, the removing conditions for the shrink-prevention ceramic sheets change. Thus, it is difficult to carry out the processing uniformly perform the processing.

-- In view of the foregoing, the

## SUMMARY OF THE INVENTION

To overcome the problems described above,

preferred embodiments of the present invention has

been devised. It is an object of the present

invention to provide a method of uniformly removing

a shrink-prevention ceramic sheet in the production

of a ceramic multi-layer substrate using the shrink
prevention ceramic sheet.

#### Disclosure of Invention

According to a preferred embodiment of the present invention, there is provided a method of producing a ceramic multi-layer substrate which includes a step of preparing a composite laminate comprising including an unfired ceramic laminate formed by lamination of laminating a plurality of substrate ceramic green sheets, and a shrink-prevention ceramic green sheet arranged on at least one of the main surfaces of the unfired ceramic laminate and being substantially incapable of being sintered at the firing temperature of the unfired

ceramic laminate, a step of firing the composite laminate at a temperature at which the unfired ceramic laminate can be fired and which is lower than the sintering temperature of the shrinkprevention ceramic green sheet, and a step of removing the shrink-prevention ceramic green sheet subjected to the firing step, from the fired composite laminate, and is characterized in thatwherein the step of removing the shrinkprevention ceramic green sheet contains includes a first removing step of spraying a liquid material (particularly, water) and compressed gas (particularly, compressed air) against the shrinkprevention ceramic green sheet on the main surface of the composite laminate subjected to the firing step, and a second removing step of spraying ceramic powder, a liquid material (particularly, water), and compressed gas (particularly, compressed air) against the main surface of the ceramic multilayer substrate subjected to the first removing step.

Preferably, the method of producing a ceramic multi-layer substrate according to this preferred embodiment of the present invention further contains includes a step of supersonic-cleaning the ceramic multilayer substrate as a third removing step subsequent succeeding after the first and second steps. Also, preferably, the method of producing a

ceramic multi-layer substrate according to this preferred embodiment of the present invention further contains includes a step of spraying a liquid material (particularly, water) and compressed gas (particularly, air) against the main surface of the ceramic multi-layer substrate as a third removing step succeeding after the first and second removing steps.

According to the method of producing a ceramic multi-layer substrate of this preferred embodiment of the present invention, mainly primarily a partportion of the shrink-prevention ceramic green sheet, which does not react with a glass component of the ceramic multi-layer substrate, can be is removed in the first removing step of spraying a liquid material and compressed gas. Thereafter, a residual material which does—is not removed in the first removing step ean be-is removed in the second removing step. Moreover, in the third removing step of supersonic-cleaning or spraying a liquid material and compressed gas, a residual material not removed in the second removing step, and also the ceramic powder sprayed in the second removing step can be are removed. Thereby, the shrink-prevention ceramic green sheet can be is uniformly removed.

## Brief Description of the Drawings

Other features, elements, steps, characteristics and advantages of the present invention will become more apparent from the following detailed description of preferred embodiments thereof with reference to the attached drawings.

## BRIEF DESCRIPTION OF THE DRAWINGS

- Fig. 1 is a schematic cross-sectional view of a ceramic multi-layer substrate according to  $\underline{a}$  preferred embodiment of the present invention.
- Fig. 2 schematically shows a process of the method of producing a ceramic multi layer substrate according to the present invention.
- Fig. 3 schematically shows a process of the step in a method of producing a ceramic multi-layer substrate according to the preferred embodiment of the present invention.
- Fig. 3 schematically shows a step in a method of producing a ceramic multi-layer substrate according to the preferred embodiment of the present invention.
- Fig. 4 is a schematic cross-sectional view of an interdigital electrode on the ceramic multi-layer substrate according to a preferred embodiment of the present invention.
- Fig. 5 is a schematic view showing the migration of an electrode on the ceramic multi-layer substrate.

## Best Mode for Carrying out the Invention

## DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

Hereinafter, the present invention will be described with reference to an embodiment preferred embodiments.

## First Preferred Embodiment

 Process of Preparing Composite Laminate and Firing the Same

As shown in Fig. 1, a composite laminate 1 is prepared by laminating a plurality of ceramic green sheets 2 for a—substrate conductive layers 3 to form an unfired ceramic laminate 4, and laminating and press-bonding shrink-prevention ceramic sheets 5 to both of the main sidessurfaces of the unfired ceramic laminate 4. In the composite laminate 1, via-conductors 6 are formed so as to connect the conductive layers 3 provided at different heights. It is to be noted than the shrink-prevention ceramic sheets 5 may be provided on one of the main sides

The substrate ceramic green sheets 2 may be prepared, e.g., by adding a binder, a plasticizer, and a solvent to ceramic powder, mixing them by means of using a ball mill, an atractor, or the likeother suitable device to form a slurry, and forming the slurry into a sheet withhaving a

thickness of about 25  $\mu m$  to about 200  $\mu m$  by a doctor blade method or the like other suitable method.

As a ceramic powder used to form s—a substrate ceramic green sheet, so-called LTCC (Low Temperature COCo-Fired Ceramic) powder may be used. For example, crystallized glass with having a crystallized temperature of about 600°C to about 1,000°C may be used, or the crystallized glass having a ceramic filler, such as alumina, zirconia, mullite, cordierite, anosite, silica, or the likeother suitable ceramic filler, added thereto may be used. Moreover, as a binder, for example, polyvinylbutyral, a methacrylic polymer, an acrylic polymer, or the likeother suitable binder may be used. As a plasticizer, derivatives of phthalic acid or the likeother suitable plasticizer may be used. As a plasticizer, alcohols, ketones, chlorine type organic solvents, or the like mayother plasticizers may also be used.

The conductive layer 3 includes a so called surface conductor layer and an internal conductor layer. The conductor layer 3 is formed by printing conductor paste containing metal powder of Ag, Cu, or the like—other suitable metal powder onto the substrate ceramic green sheet 2. Moreover, the viaconductor 6 provided for the composite laminate 1 is formed by filling the conductor paste into a via-

hole formed in the substrate ceramic green sheets 2.

The shrink-prevention ceramic green sheet 5 is prepared byusing the same production method as that for the substrate ceramic green sheet 2. However, the sintering temperature is higher than the temperature at which of the substrate ceramic green sheets 2. For example, in the case in which where ceramic powder having a sintering temperature of up to about 1,100°C is used for the substrate ceramic green sheets 2, alumina, zirconium oxide, aluminum nitride, boron nitride, mullite, magnesium oxide, silicon carbide, or the like other suitable material may be used. Suitably, the The average particle sizes of these ceramic powders are in the range of about 0.5  $\mu m$  to about 4  $\mu m$  . If the particle size is too large, the shrink-controlling force for the substrate ceramic sheets becomes weak in some cases.is reduced. Moreover, the surface of the formed ceramic multi-layer substrate becomes—is rough in some casesinstances.

In press-bonding the substrate ceramic green sheets 5 to both of the main surfaces of the unfired ceramic laminate 4, the pressure applied for the press-bonding is suitablypreferably in the range of about 10 MPa to about 200 MPa, and the temperature is suitablypreferably in the range of about 40°C to about 90°C.

Thereafter, the composite laminate 1 is fired. Thus, the ceramic multi-layer substrate having the shrink-prevention ceramic green sheets 5 formed on both of the main surfaces is prepared. In this case, the The sintering temperature is required to must be a temperature at which the unfired ceramic laminate 4, that is, the substrate ceramic green sheets are sintered and which is <del>lower</del>less than the sintering temperature of the shrink-prevention ceramic green sheets 5. That is, the shrink-prevention ceramic green sheets are not sintered at the firing. That is, sincewhen fired. Since the shrink-prevention ceramic green sheets are not shrunk during the firing, the shrink of the ceramic laminate in the plane direction is suppressed. Probably, this This is because the glass component <del>oozed</del>which oozes out from the ceramic laminate at the firing and the shrink-prevention ceramic green sheets react with each other, and thus, the reaction layer is formed at the interface. Accordingly, the positional accuracy of the conductor patterns provided on the ceramic laminate can be is maintained high when it is subjected to the firing processing, and also, the disconnection thereof is suppressed prevented.

#### 2. First Removing Step

After the composite laminate is fired, a liquid

material, together with compressed air, is sprayed against the shrink-prevention ceramic green sheets. At this time, the organic component, such as a binder or the like, has been fired out and removed from the shrink-prevention ceramic green sheets. Thus, the green sheets are in the porous state. a liquid material, an acidic aqueous solution, an alkali solution, an organic solvent, or the like other suitable liquid material may be used. Particularly, water is preferable from the viewpoints of the property with respect to the environments and the cost-performance. Moreover, as compressed gas, nitrogen gas or the like other suitable gas may be used. Particularly, compressed air is desirable preferable from the viewpoint of the cost--performance.

As a method of spraying a liquid material together with compressed gas, e.g., a method using a plasto-nozzle as shown in Fig. 2 is exemplified.used as an example. In particular, a ceramic multi-layer substrate 24, which has been subjected to a firing process, is placed on a stand 7. Thereafter, a sealed material (here, water) is sprayed against a shrink-prevention ceramic green sheet 25 formed on one of the main surfaces of the ceramic multi-layer substrate 24, while the sealed material 8 is accelerated with compressed gas 9. In this case, a

A mixture 11 of the water 8 and the compressed gas 9 is discharged through a nozzle 10, which is a discharge port of the plasto-nozzle. Then, the mixture 11 is continuously sprayed while the nozzle 10 is sequentially scanned in a direction A shown by the arrow in Figure. 2.

In-this case, the The pressure of the compressed gas is desirably preferably in the range of about 147 kPa to about 539 kPa. If the processing is carried outperformed at a pressure of less than about 147 kPa, the removing capacity for the shrink-prevention ceramic green sheet is inferior, since the spraying pressure is excessively low, and the production efficiency decreases. On the other hand, if the processing is carried outperformed at a pressure of more than about 539 kPa, the degradation of the nozzle 10 is accelerated due to the pressure. Moreover, the consumption of the compressed gas 9 increases, which causes a highincreased running cost and also may damage the ceramic multi-layer substrate 24. Here, the pressure of compresses gas means a pressure thereof in a—the piping before the spraying being sprayed.

By carrying out the first removing step, a partportion of the shrink-prevention ceramic green sheet 25 which has not reacted with a glass component of the ceramic multi-layer substrate 24 is removed. As a result, the reaction layer formed by the reaction of the shrink-prevention ceramic green sheet 25 with the glass component of the ceramic multi-layer substrate 24 remains. Also, in some cases, the non-reacted part portion of the shrink-prevention ceramic green sheet 25, which does not react with the glass component of the ceramic multi-layer substrate 24, is not removed by the first removing step—and remains. Fig. 2—exaggeratedly shows the—a state in which the shrink-prevention ceramic green sheet 25 is partially removed, while no residual material of the shrink-prevention ceramic green sheet 25 is shown in the drawing.

Powder recovered in the first removing step essentially consists of the ceramic powder generated from the shrink-prevention ceramic green sheet.

Therefore, the powder can be efficiently recovered and re-used as ceramic powder particularly for use in a shrink-prevention ceramic green sheet.

## 3. Second Removing Step

Thereafter, ceramic powder and a liquid material are sprayed, together with compressed gas, against both of the main surfaces of the ceramic multi-layer substrate which has have been subjected to the first removing step. In this method, for example, they are sprayed, using a plasto-nozzle in a manner

similar to that described with respect to the first removing step. In Fig. 2, as a sealed material 8, a mixture of ceramic powder and water is injected.

The ceramic powder, water, and compressed gas are discharged through the nozzle 10. Here, as a liquid material, an acidic aqueous solution, an alkali aqueous solution, an organic solvent, and so forth or other suitable liquid materials may be also used. From the viewpoints of the property with respect to the environments and the cost—performance, particularly, water is desirable. Preferable. As compressed gas, nitrogen gas may be used. From the viewpoint of the cost—performance, particularly, compressed air is desirable preferable.

In this case, the The pressure of the compressed gas is desirably preferably in the range of about 98 kPa to about 343 kPa. If the processing is carried outperformed at a pressure of less than about 98 kPa, the removing capacity for the shrink-prevention ceramic green sheets is inferior, since the spraying pressure is excessively low, and the production efficiency decreases. If the processing is carried outperformed at a pressure of more than about 343 kPa, cracks are readylikely to occur at the interfaces between the surface conductor layers and the ceramic multi-layer substrate. Thus, the bonding strength between the conductor layers and

the composite laminate is reduced. Thus, inconveniently, Therefore, peeling of the conductor layers in the plating step or the like is likely to occur in some cases. In this case, it is desirable that the pressure is preferably smaller than that in the first removing step. That is, in the second removing step, the ceramic power is used as abrasive grains. Thus, if the pressure of the compressed air is higher than that in the first removing step, the surface property of the ceramic multi-layer substrate, particularly, the surface properties of the conductor layers of the ceramic multi-layer substrate will be deteriorated in some cases.

Desirably Preferably, the average particle size of the ceramic powder for spraying is in the range in the range of about 9.5 μm to about 40 μm. If ceramic powder having an average particle size of less than about 9.5 μm is used, the removing capacity for the shrink-prevention ceramic green sheets is inferior, and the production efficiency is reduced in some cases. On the other hand, if ceramic powder having an average particle size of more than about 40 μm is used, the collision force is large at increased during spraying, sosuch that cracks are readylikely to occur at the interfaces between the conductor layers and the composite laminate. Thus, the bonding strength is reduced,

and inconveniently, the conductor layers are peeled and so forth in some cases likely to peel in the plating step. Moreover, problems occur in that the processing of portions where at which the intervals between wirings are small is ready likely to be irregular and so forth, since the particle size is large.

When the second removing step is earried out, the performed, most of a the trace amount of residual materials on both of the surfaces of the ceramic multi-layer substrate, which have not been removed in the first removing step, are removed, due to the physical action of the liquid material (particularly, water), the ceramic powder, and the compressed gas (particularly, compressed air). The ceramic powder recovered in the second removing step essentially consists of the ceramic powder sprayed together with water. Therefore, the ceramic powder can be efficiently recovered and re-used particularly as ceramic powder to be sprayed.

## 4. Third Removing Step

Thereafter, the ceramic multi-layer substrate which has been subjected to the first and second removing steps is cleaned by a supersonic wave method.

In this step, as shown in Fig. 3, a cleaning

liquid 13 is placed in a cleaning tank 12. The ceramic multi-layer substrate 24 as a material to be eleaned—is placed into a cleaning basket 14 disposed in the cleaning tank 12. Thus, supersonic waves are irradiated into the cleaning liquid 13 by use of—a supersonic wave vibrator 16—connected—to—a supersonic wave vibrator 16. Examples of the cleaning liquid include a methylene chloride aqueous solution, a trichloroethylene aqueous solution, and so forthother suitable cleaning liquids.

In the case, it It is desirable preferable that the ceramic multi-layer substrate 24 is set in the cleaning basket 14 so as to stand upright. In this step, residual materials which have not been removed in the first and second removing steps, and the ceramic powder sprayed in the second removing step and remaining which remains on the surface are removed.

Desirably Preferably, the vibrator frequency employedused in the supersonic cleaning is in the range of about 40 tokHz to about 100 kHz. If the processing is earried outperformed at about 40 KHz or less, the cavitation force is high, so increases, such that the substrate is causes to swingswings considerably during the processing. In the case of where a substrate having has a small thickness, the substrate is cracked in some cases.may crack.

Conductor layers which position located in the vicinity of the vibrating portion of the vibrator may be broken. Moreover, in the case in which when the cavitation force is high, the removing capacity for the ceramic powder placed into the porous portions of the ceramic layer and the conductor layers is low. This will cause problems such as irregular plating,—and abnormal deposition,—and—so forth. If the processing is carried out atat about 100 KHz or higher, the cavitation force is extremely low, and the removing effect on the ceramic powder (sprayed ceramic powder) remaining on the surface of the ceramic multi-layer substrate is reduced. This results in low production efficiency—in—some—cases.

Desirably Preferably, the output per unit area of the supersonic vibrator is in the range of about 0.2 to W/cm² to about 2.0 W/cm². If the processing is carried outperformed at less than about 0.2 W/cm², the removing effect on the ceramic powder (sprayed ceramic powder) remaining on the surface of the ceramic multi-layer substrate on the like decreases, and the production efficiency is reduced in some cases. On the other hand, if the processing is carried out atat about 2.0 W/cm² or higher, the substrate is caused to swingswings considerably during the processing. In the case of where a substrate havinghas a small thickness, it

Conductor layers which position located in the vicinity of the vibrating portion of the vibrator are may be significantly damaged and broken in some cases. Moreover, the removing capacity for the ceramic powder placed into the porous portions of the ceramic layer and the conductor layers is low. This will cause problems such as irregular plating and abnormal deposition, and so forth.

When the supersonic cleaning is carried outperformed, residual materials which have not been removed in the second removing step, and the ceramic powder which has been sprayed in the second removing step, are removed from the main surfaces of the ceramic multi-layer substrate 24, due toby the physical action of the cavitation and the chemical action of a cleaning agent.

# Second Preferred Embodiment

Similarly to the above-described first preferred embodiment, the composite laminate having the shrink-prevention ceramic green sheets formed on both of the main surfaces is prepared, and is subjected to the first and second removing steps.

## 5. Third Removing Step

A liquid material is sprayed, together with

compressed gas, against the ceramic multi-layer substrate which has been subjected to the first and second removing steps. In this case, e.g., the method using a plasto-nozzle as empoyed insimilar to the first removing step according to the first preferred embodiment may be used. Here, as a liquid material, an acidic aqueous solution, an alkali aqueous solution, an organic solvent, and soforthother suitable liquid materials may be also used. From the viewpoints of the property wit respect to the environments environmental characteristics and the cost performance, particularly, water is desirable.preferable. compressed gas, nitrogen gas may be used. From the viewpoint of the cost performance, particularly, compressed air is desirable preferable.

In this case, the The pressure of the compressed gas is desirably preferably in the range of about 147 tokPa to about 539 kPa. If the processing is carried outperformed at a pressure of less than about 147 kPa, the removing capacity for the shrink-prevention ceramic green sheets is inferior, since the spraying pressure is excessively low, and the production efficiency decreases. On the other hand, if the processing is carried out at a pressure of more than about 539 kPa, the degradation of the nozzle 10 is accelerated due to the pressure.

Moreover, the consumption of the compressed gas 9 increases, so such that the running cost becomes highincreases, and the ceramic multi-layer substrate 24 is may be damaged in some cases. In this case, it is desirable. It is preferable that the pressure is higher greater than that in the second removing step. Thus, if the pressure of the compressed air is lower less than that in the second removing step, it will be difficult to remove the ceramic powder (sprayed ceramic powder) intruding into the surface of the ceramic multi-layer substrate in the second removing step.

#### EXAMPLE

Hereinafter, the present invention will be described with reference to specific examples. EXAMPLE 1.

First, 15 parts by weight of polyvinylbutyral, 40 parts by weight of isopropylalcohol, and 20 parts by weight of trol were added to 100 parts by weight of mixed powder formed by mixing crystallized glass powder containing  $SiO_2$ ,  $Al_2O_3$ ,  $B_2O_3$ , and CaO with alumina powder at an equi-percents by weight, and mixed with a ball mill for about 24 hours to form a slurry. The slurry was formed into a ceramic sheet with a thickness of about 120  $\mu$ m. Thus, a ceramic green sheet for a substrate was formed.

Subsequently, Ag paste was screen-printed thereon in a predetermined position thereof.location. The ceramic green sheets for a substrate were coated as described above, and an interdigital electrode was formed. As shown in Fig. 4, an interdigital electrode 17 comprises includes first electrode fingers 18a formeddisposed on a first terminal 18, and second electrode fingers 19a on a second terminal 19 which are formedarranged in opposition to each other on the substrate ceramic green sheet 2. The widths of the first electrode fingers 18a and the second electrode fingers 19a are preferably about 100  $\mu$ m, and the intervals between the first electrode fingers 19a are about 100  $\mu$ m.

Then, 15 parts by weight of polyvinylbutyral, 40 parts by weight of isopropylalcohol, and 20 parts by weight of trol were added to 100 parts by weight of alumina powder, and mixed with a ball mill for 24 hours to form a slurry. The slurry was formed into a ceramic sheet with a thickness of about 120  $\mu$ m by a doctor blade method. Thus, a shrink-prevention ceramic green sheet was formed.

Then, six substrate ceramic green sheets were laminated, and also, the shrink-prevention ceramic green sheets were laminated to both of the main surfaces of the six substrate ceramic green sheets,

respectively. Then, the sheets were press-bonded at a pressure of <u>about</u> 150 MPa and a temperature of <u>about</u> 60°C. Thus, a composite laminate was formed.

Thereafter, the composite laminate was placed on a tray made of an alumina plate with a flatness degree of up to <u>about</u> 0.05% and a porosity of <u>about</u> 70%, and heated at <u>about</u> 600°C for <u>about</u> 3 hours, and thereafter, pressed at a temperature of <u>about</u> 900°C for <u>about</u> 1 hour. Thus, the substrate ceramic green sheets were sintered.

Subsequently, in the first removing step, water together with compressed air having the respective pressures in the range of <u>about 147 kPa</u> to <u>about 539 kPa</u> shown in Table 1, was sprayed for <u>about 120 seconds</u> against the shrink-prevention ceramic green sheets provided on both of the main surfaces of the ceramic multi-layer substrate.

Thereafter, in the second removing step, water and alumina powders with the respective average particle sizes in the range of about 9.5 µm to about 40 µm shown in Table 1, together with compressed air in the range of about 98 kPa to about 343 kPa were sprayed for about 120 seconds against residual materials on the ceramic multi-layer substrate after the first removing step.

Subsequently, in the third removing step, supersonic cleaning was carried out at a supersonic

vibrator frequency of about 40 kHz to about 100 kHz and an output per unit supersonic vibrator of about 0.2  $\text{W/cm}^2$  to about 2.0  $\text{W/cm}^2$  for about 300 seconds.

As a result of the above-described steps, the ceramic multi-layer substrates of Samples 1 to 8 were formed.

#### Comparative Example 1

A composite laminate was formed and fired under the same preparation conditions as those in First Example. The shrink-prevention ceramic green sheets were removed from the composite laminate, not subjected to the second removing step. In particular, in the first removing step, water and compressed air with about 539 kPa were sprayed for about 120 seconds against the shrink-prevention ceramic green sheets provided on both of the main surfaces of the ceramic multi-layer substrate. Thereafter, in the third removing step, supersonic cleaning was carried out at a supersonic vibrator frequency of about 40 kHz, an output per unit area of a supersonic vibrator of about 0.2/cm2 for about 300 seconds. As a result of the above-described steps, the ceramic multi-layer substrate of First Comparative Example was produced.

#### Second Comparative Example 2

A composite laminate was formed and fired in the

same conditions as those in Example First.1. samples were prepared. For these These twenty samples not were subjected to the first removing step, the second and third removing steps were carried out to remove the shrink-prevention ceramic green sheets, but not the first removing step. the second removing step, water and alumina powder with an average particle size of about 9.5  $\mu$ m, together with compressed air with about 98 kPa, were sprayed for about 120 seconds against both of the main surfaces of the ceramic multi-layer substrate. Thereafter, in the third removing step, the twenty substrates were supersonic-cleaned at a supersonic vibrator frequency of about 40 kHz and an output per unit area of a supersonic vibrator of about 0.2/cm<sup>2</sup> for about 300 seconds. For the twenty samples, the second and third removing steps were carriedperformed sequentially, out one by one from the first sample to the twentieth sample. these steps, a ceramic multi-layer substrate of comparative example 2a (first sample) and a ceramic multi-layer substrate of comparative example 2b (second sample) were prepared.

Regarding the respective samples (samples Nos. 1 to 8, comparative example 1, and comparative examples 2a and 2b) prepared in EXAMPLE 1, COMPARATIVE EXAMPLE 1, and COMPARATIVE EXAMPLE 2,

the appearances were examined, and the results were shown in Table 1, where a circular mark represents "no irregular-removal", and a cross mark does "irregular removal".

Thereafter, a palladium catalyst was givenapplied to the respective samples of the ceramic multi-layer substrates, and then, the cleaning was earried out, soperformed, such that nuclei of the palladium catalyst were formed on the interdigital electrode portion. Thereafter, electroless nickel plating was earried out performed. Thus, nickel plating was performed on the interdigital electrode portion. A voltage of about 50 V was applied to the interdigital electrode portion under the conditions of about 85°C and about 85%RH for about 1,000 hours. Then, the insulation resistance was measured. Table 1 shows the measuring results.

Table 1

Sample Number	First removing step	Second removing step		Third removing step		Annograna	Insulation resistanc
	Pressure (kPa)	Average particle size (µm)	Pressure (kPa)	Frequenc y (KHz)	Output (W/cm²)	Appearanc e of substrate	e LogIR (Ω)
No. 1	147	9.5	98	40	0.2	0	≥9
No. 2	539	9.5	98	40	0.2	0	≥9
No. 3	147	9.5	343	40	0.2	0	≥9
No. 4	147	40	98	40	0.2	0	≥9
No. 5	147	40	343	40	0.2	0	≥9

No. 6	147	9.5	98	40	2.0	0	≥9
No. 7	147	9.5	98	100	0.2	0	≥9
No. 8	147	9.5	98	100	1.0	0	≥9
Comparati ve example 1	539	Not processed		40	0.2	×	≤5
Comparati ve example 2a	Not processed (first sample)	9.5	98	40	0.2	0	≥9
Comparati ve example 2b	Not processed (twentieth sample)	9.5	98	40	0.2	×	≤5

#### EXAMPLE 2

Similarly to EXAMPLE 1, the ceramic multi-layer substrate having the shrink-prevention ceramic green sheets formed on both of the main surfaces was prepared. The first and second removing steps were carried outperformed under the conditions shown in Table 2.

Thereafter, in the third removing step, water, togethercombined with compressed air in the range of about 147 kPa to about 539 kPa, was sprayed for about 120 seconds against the ceramic multi-layer substrate subjected to the first and second removing steps.

As a result of these steps, the ceramic multilayer substrates of Samples Nos. 9 to 14 shown in Table 2 were prepared.

#### COMPARATIVE EXAMPLE 3

A composite laminate was formed and fired under the same preparation conditions as those described in EXAMPLE 2. The shrink-prevention ceramic green sheets were removed from the composite laminate, while not being subjected to the second removing step. In particular, in the first removing step, water and compressed air with about 539 kPa were sprayed for about 120 seconds against the shrinkprevention ceramic green sheets provided on both of the main surfaces of the ceramic multi-layer substrate. Thereafter, in the third removing step, water and compressed air in the range of about 147 kPa to about 539 kPa were sprayed for about 120 seconds. As a result of the above-described steps, the ceramic multi-layer substrate of Third Comparative Example was produced.

#### COMPARATIVE EXAMPLE 4

A composite laminate was formed and fired inunder the same conditions as those described in EXAMPLE 2. Thus, twenty samples were prepared. For these twenty samples which were not subjected to the first removing step, the second and third removing steps were carried out performed to remove the shrink-prevention ceramic green sheets. In the second removing step, water and alumina powder with an average particle size of about 9.5 μm, together

with compressed air withat about 98 kPa, were sprayed for about 120 seconds against both of the main surfaces of the shrink-prevention ceramic green sheets provided on both of the main surfaces of the ceramic multi-layer substrate. Thereafter, in the third removing step, water, together with compressed air in the range of about 147 kPa to about 539 kPa, was sprayed for about 120 seconds against the twenty samples. For the twenty samples, the second and third removing steps were <del>carried out</del>performed sequentially, one by one from the first sample to the twentieth sample. After these steps, a ceramic multi-layer substrate as comparative example 4a (first sample) and a ceramic multi-layer substrate as comparative example 4b (second sample) were prepared.

Regarding The appearance of the respective samples (samples Nos. 9 to 14, comparative example 3, and comparative examples 4a and 4b) prepared in EXAMPLE 2, COMPARATIVE EXAMPLE 3, and COMPARATIVE EXAMPLE 4, the appearances were examined, and the results were shown in Table 1, where a circular mark represents "no irregular-removal", and a cross mark does "irregular removal".

Thereafter, a palladium catalyst was given applied to the respective samples of the ceramic multi-layer substrates, and then, the

cleaning was carried out, soperformed, such that nuclei of the palladium catalyst were formed on the interdigital electrode portion. Thereafter, electroless nickel plating was carried out.performed. Thus, nickel plating was performed on the interdigital electrode portion. A voltage of about 50 V was applied to the interdigital electrode portion under the conditions of about 85°C and about 85%RH for about 1000 hours. Then, the insulation resistance was measured. Table 2 shows the measuring results.

Table 2

Sample Number	First removing step		emoving ep	Third removing step	Appearanc	Insulation resistance LogIR (Ω)
	Pressure (kPa)	particle size (μm)	Pressure (kPa)	Pressure (kPa)	e of substrate	
No. 9	147	9.5	98	147	0	≥9
No. 10	539	9.5	98	147	0	≥9
No. 11	147 .	9.5	343	147	0	≥9
No. 12	147	40	98	147	0	≥9
No. 13	147	40	343	147	0	≥9
No. 14	147 .	9.5	98	539	0	≥9
Comparativ e example 3	539	Not processed		539	×	≤5
Comparativ e example 4a	Not processed (first sample)	9.5	98	539	0	≥9
Comparativ e example 4b	Not processed (twentieth sample)	9.5	98	539	×	≤5

As described above, in this example, the shrink-

prevention ceramic green sheets <u>could bewere</u>
uniformly removed, and <u>moreover</u>, <u>the goodoutstanding</u>
insulation resistance <u>could be</u> was maintained.

On the other hand, in the comparative example 1, the appearance showed the irregular removal was clearly shown, and the insulation resistance represented by LogIR was not more than about 5.

In the comparative example 2, the appearance of the—first ceramic multi-layer substrate showed had no irregular removal, while that of the second ceramic multi-layer substrate showedhad some irregular removal. Regarding the insulation resistance, the LogIR of the first ceramic multi-layer substrate was not less than about 9, while that of the twentieth ceramic multi-layer substrate was not more than about 5. That is, it has been revealed that the repetition of the second removing step causes the removing capacity for the shrink-prevention ceramic green sheets to be greatly reduced.

In the comparative example 3, the appearance showed—irregular removal as clearly shown, and the insulation resistance represented by LogIR was not more than about 5.

In the comparative example 4, the appearance of the first ceramic multi-layer substrate showed had no irregular removal, while that of the twentieth

ceramic multi-layer substrate showedhad some irregular removal. Regarding the insulation resistance, the first ceramic multi-layer substrate had a LogIR of not less than about 9, while the twentieth ceramic multi-layer substrate has a LogIR of not more than about 5. That is, it has been revealed that the repetition of the second removing step causes the removing capacity for the shrink-prevention ceramic green sheets to be greatly reduced.

The reason for the reduction in insulation resistance of the twentieth ceramic multi-layer substrates of the comparative examples 1 and 3 and the comparative examples 2 and 4 is as follows.

That is, as described below. As shown in Fig. 5, a residual material 20 made fromof the shrink-prevention ceramic green sheets exists on surfaces of a ceramic multi-layer substrate 34. Thus, a partportion of the end surfaces 17a of the electrode 17 of the interdigital electrode is covered with the reamingresidual material. The residual material 20 is porous. Thus, Migration migration of Ag occurs in the potion portion where no plating is performed, in the direction shown by an arrow, so such that the insulation resistance is reduced.

## Industrial Applicability

As described above, according to the method of producing a ceramic multi-layer substrate according to preferred embodiments of the present invention, produces ceramic multi-layer substrates having a high dimensional accuracy can be suitably are produced at a with high efficiency.

with respect to preferred embodiments, it will be apparent to those skilled in the art that the disclosed invention may be modified in numerous ways and may assume many embodiments other than those specifically set out and described above.

Accordingly, it is intended by the appended claims to cover all modifications of the present invention that fall within the true spirit and scope of the invention.